A CONTRIBUTION TO THE FISH OIL THEORY OF THE ORIGIN OF PETROLEUM FROM A STUDY OF BRINE WATERS FROM WIDELY DIFFERENT OIL FIELDS IN JAPAN.

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Introduction. The widely known fact that brine water and petroleum occur simultaneously in almost all of the oil fields in the world gives an interesting but unsolvable problem to scientists, but gives a cause of great annoyance to the workers of petroleum industry. Though no apparent relationship may exist between brine water and petroleum, yet their mysterious intimacy in nature tells us some intrinsic relation between them, and if this would be brought to light, the mystery concerning the origin of petroleum would probably be disclosed, and in consequence we would have some important clue which must be of great service to the discovery of petroleum deposit. This thought led some of the earlier petroleum geologists and chemists to their earnest studies of brines from different oil fields and to give rise to several theories of their origins. Prior to any other investigators, Hunt[(1)] in 1875 suggested from his study of the brine water from Ontario that it comes from the ancient sea water embedded simultaneously with the formation of the petroleum deposit. In 1905 Lane[(2)] extended this idea more generally and published a well-known theory named the connate or fossil water theory, which states that the brine water from an oil field is no other than the ancient sea water secluded from the outer side and preserved in strata of earth. On the other hand, Washburne[(3)] and Richardson[(4)] have somewhat different opinions that the salts contained in brine water have not their origin in those of the sea water as the forementioned investigators believed, but in some other salts which might come from easily soluble basic magmas or salt deposits. These opinions, however, do not seem to be adequate at least in Japan whose geological conditions differ from those in foreign countries. It may therefore be mentioned here that the acceptable theories at present are those which stand on sea water, though in particulars they are differently modified by many investigators.

Analyses of the Japanese Brine and the Discussion of Their Results. The present authors who have had much interest in the co-existence of brine and petroleum for a long while had an opportunity to get the brines from widely different oil fields in Japan for which there have been little known in the literature. The brines taken for the analyses are those from the oil fields in Akita and Niigata Prefectures and that from the natural gas field with an oil indication in Chiba Prefecture. The results of the analyses are shown in Table 1, where the composition is given in molar ratios taking the sodium as 100%. Column 8 shows the absolute concentration of the sodium (grams per litre) in each brine for reference. Parallel to Table 1, Table 2 shows the composition of the sea waters from several places in the world which are all quoted from other authors. As seen from Table 1 and Table 2, it is clearly understood that the composition of sea waters give as already considered quite concordant values to each other independent of their occurring places, and that of brines are also, though not fairly concordant, yet roughly convergent to certain definite ratios in spite of the diversity in their geological distribution. Remarkable similarities are found between the composition of the sea water and that of the Japanese brine water in the ratios of the sodium, potassium and calcium, but far less quantities are observed in the magnesium and sulphate in the case of the brine, especially the latter being a trace or naught. The chlorine also seems to have somewhat lower values in the brine. As for the minute content of the sulphate in the brine from oil fields which is true not only for Japan, but also for any other countries, many explanations have been proposed by foreign investigators: Höfer[9] and Rogers[10] by whom the former's theory was supported, proposed that the lack of the sulphate may be due to the interaction between brine water and petroleum in the following chemical change:

$$SO_4^{--} + CH_4 \rightarrow CO_3^{--} + H_2O + H_2S,$$
$$\text{or} \rightarrow S^{--} + 2H_2O + CO_2$$

The idea was rejected by Palmer[11] who stated that these reactions, being endothermic, can not be well explained unless some favorable con-

Table 1. Composition of Japanese brines.

Brines from	Na	K	Ca	Mg	Cl	SO_4	Na	Authors
1 Innai, Akita Pref.	100	2.16	0.82	1.67	105.2	0.01	4.69	Ishikawa & Baba
2 Michikawa, Akita Pref.	100	0.21	5.34	5.31	110.8	0.01	6.00	,,
3 Niizu, Niigata Pref. (Mean of 2 samples)	100	2.32 (3.02 1.62)	1.16 (1.63 0.68)	0.73 (1.15 0.30)	79.1 (82.6 75.6)	0.01 (0.02 0.00)	4.40 (4.76 4.03)	,,
4 Nishiyama, Niigata Pref. (Mean of 4 samples	100	0.32 (0.35 0.27)	1.91 (2.62 1.57)	0.49 (0.61 0.40)	95.1 (101.7 90.7)	0.00	6.72 (7.28 6.16)	,,
5 Ôtaki, Chiba Pref.	100	1.17	0.74	1.14	102.3	—	9.75	,,
6 Katsurane, Akita Pref. (Mean of 6 samples)	100	2.42 (4.27 1.09)	1.88 (6.70 0.31)	0.96 (2.51 0.30)	104.2 (117.4 79.1)	0.12 (0.36 0.02)	4.16 (8.05 1.02)	Mikawa[5]
7 Ôtaki, Chiba Pref. (Mean of 6 samples)	100	0.87 (1.96 0.62)	0.77 (1.46 0.23)	2.86 (4.53 0.92)	89.0 (109.5 54.6)	—	2.67 (6.40 0.88)	Ôhashi[6]

(1) The Asahi Oil Co.'s well, 381.5–385.5 m. deep, at Innai-mura, Yuri-gun, Akita Prefecture.

(2) The Asahi Oil Co.'s well, 162 m. deep, at Michikawa-mura, Yuri-gun, Akita Prefecture.

(3) The Asahi Oil Co.'s well, 93 m. deep, of Aozawa, Take, Niizu-cho, Niigata Prefecture and the same Co.'s well, 270 m. deep, at Okubozawa, Take, Niizu-cho, Niigata Prefecture.

(4) The Nippon Oil Co.'s wells, No. 33, 880 m. deep; No. 24, 771 m. deep; No. 60, 1200 m. deep; and No. 35, 1228 m. deep, at Taka-machi, Kariha-mura, Kariha-gun, Niigata Prefecture.

(5) The Ôtaki Natural Gas Co.'s No. 2 well, at Oyamatsu, Fusamoto-mura, Isumi-gun, Chiba Prefecture.

Parenthesized figures show the highest and lowest values respectively.

Table 2. Composition of sea waters.

Seas	Na	K	Ca	Mg	Cl	SO_4	Br	Na
Mean of 77 samples collected during the voyage of Challenger	100	2.12	2.25	11.50	117.2	6.02	0.176	10.77
Between Cape of Good Hope and England (Mean of 22 samples)	100	2.07	2.36	12.17	118.2	6.26	0.170	10.99
Near Carthage, Mediterranean	100	2.11	2.38	11.33	118.6	6.10	0.171	11.84
Black Sea (Mean of 6 samples)	100	2.24	2.66	11.61	117.3	5.87	0.170	6.17
Red Sea (Mean of 4 samples)	100	2.06	2.31	11.39	119.7	5.91	0.171	16.58
Genkai-Nada, Sea of Japan[7]	100	2.18	2.35	11.33	114.9	5.96	—	10.11
Inland Sea, Japan[8]	100	2.02	2.03	11.76	117.4	6.12	—	10.12

Table 3.

Composition of animal bloods (sera or plasmas).

Animals	Na	K	Ca	Mg	Cl	SO_4	Authors
Medusa : Aurelia flavidula	100	3.05	2.37	10.81	120.3	3.79	Macallum(14)
Horseshoe crab : Limulus polyphemus	100	3.31	2.33	10.59	121.3	3.83	,,
Lobster : Homarus americanus	100	2.19	2.78	1.63	111.0	1.92	,,
Sharks : Acanthias vulgaris	100	2.71	1.56	2.33	107.5	—	,,
Mustelus canis	100	1.63	1.89	1.07	87.8	0.96	Smith(15)
,,	100	2.07	2.15	1.11	85.2	0.66	,,
Carcharias littoralis	100	2.13	1.94	0.89	88.4	0.12	,,
,,	100	1.96	2.18	0.84	87.6	0.25	,,
Rays : Raja stabuloforis	100	1.91	1.45	0.65	88.6	trace	,,
,,	100	1.91	1.23	1.02	97.5	trace	,,
,,	100	2.04	1.49	1.37	92.2	0.20	,,
,,	100	—	1.69	1.31	99.6	trace	,,
Raja diaphenes	100	2.87	2.15	1.48	95.8	1.31	,,
Fishes : Lophius piscatorius	100	3.48	1.74	0.50	95.0	0.60	,,
,,	100	3.76	2.10	8.07	112.4	1.99	,,
,,	100	4.39	1.02	0.68	81.0	1.76	,,
,,	100	3.42	1.09	—	89.1	1.14	,,
Gadus callarias	100	2.72	2.78	2.11	87.8	—	,,
,,	100	5.59	2.26	1.33	97.1	—	Macallum(14)
Pollachius virlus	100	2.55	1.78	1.38	89.4	—	,,
Amiataus calva	100	1.29	4.54	0.30	90.9	1.06	Smith(15)
,,	100	1.73	3.46	0.30	89.5	2.26	,,
Lepidosteus osseus	100	1.93	4.36	0.21	84.3	1.71	,,
Turtles : Chrysemys marginata	100	2.50	5.08	6.58	67.7	—	,,
,,	100	2.42	3.22	1.07	59.9	0.54	,,
Graptemys geographia	100	1.94	2.74	0.40	69.8	0.32	,,

Table 3.—(*Concluded*).

Animals	Na	K	Ca	Mg	Cl	SO_4	Authors
Turtles : Emys blandingii	100	3.00	2.54	1.62	70.0	1.00	Smith(15)
Pseudemys elegans	100	2.62	3.33	3.49	64.1	0.08	,,
Chelydra serpentina	100	2.48	4.61	3.19	38.7	0.21	,,
,,	100	2.22	3.22	1.43	54.4	0.21	,,
Caretta kempi	100	4.05	3.19	0.86	66.4	0.18	,,
Caretta caretta	100	4.44	2.05	1.92	72.7	—	,,
Dog	100	3.97	1.43	0.83	84.2	—	Abderhalden(16)
,,	100	3.49	1.47	0.72	82.4	—	,,
Cat	100	3.89	1.37	0.75	82.0	—	,,
,,	100	3.94	1.24	0.87	66.1	—	Baumann & Kurland(17)
Rabbit	100	3.84	1.45	0.80	76.0	—	Abderhalden(16)
Ox	100	4.00	1.42	0.75	74.6	—	,,
Pig	100	4.18	1.59	0.74	74.5	—	,,
Horse	100	3.84	1.41	0.81	73.2	—	,,
Man	100	3.42	1.71	0.86	69.2	1.37	Kramer & Tisdall(18)

dition of thermochemical or geochemical action should occur.

Mills and Wells (12) considered the diminution of the sulphate in the brine to be due to the adsorption of it by some colloidal particles charged with positive electricity.

Recently Bastin (13) elucidated the same phenomenon by the action of micro-organisms whose metabolism necessarily consumes the sulphate in the brine.

Although the above explanations may prove the natural tendency of the unusual diminution of the sulphate in the brine, they can not interpretate the variation of the elements whose contents differ much from the contents in the sea water. In fact, little attention has been hitherto made in regard to the comparatively small content of the magnesium in the brine.

Taking the substantial differences between the sea water and the Japanese brine in consideration, the authors hesitate to accept the connate water theory which necessitates the introduction of some other assumptions of different viewpoints in order to elucidate the fact. It must be remembered that a question whether or no the ancient sea water was possessed of the same composition as the contemporary one remains unsettled. This question is no doubt of interest from the geological and biological standpoints, but also of importance from the authors' viewpoint at the same time, as will be shown later on.

In 1910 Macallum[14] put forward an interesting suggestion to this problem. He proposed, having analysed the inorganic constituents in the bloods of several animals (1) that the inorganic blood composition of an animal of a certain genus is entirely invarient ever since that genus took the first appearance on the earth and (2) that the inorganic blood composition of an animal created as an ancester of a genus must be identical with that of the surrounding sea water at that time in order to be suited to environments. Table 3 shows the inorganic composition of blood sera or plasmas of various kinds of animals in the same expressions as taken in Table 1 and Table 2. The comparison of Table 3 with Table 2 which shows the composition of sea waters brings us that there exists some dissimilarity between them except the jelly-fish and horseshoe crab. The exceptional cases seen in the jelly-fish and horseshoe crab were regarded by Macallum as such, with a reason that they have incomplete excretory organs which admit sea water to pass through their bodies freely.

In the authors' opinion, however, his conception for the sea water of olden times seems to be wrong from the consideration of the paleontological fact that the jelly-fish appeared already in Cambrian formation and the horseshoe crab somewhat later but its larva quite resembles to the trilobite which appeared likewise in Cambrian, and it is highly probable to assume that the inorganic blood composition of these animals may indicate no other than the composition of the ancient sea water, and that that of the other animals which appeared far later than these have no such composition as that of the ancient sea water on account of their gradual development of organs.

It is also plausible from a geological standpoint of view that the composition of the ancient sea water did not differ so much from the to-day's so far as it is concerned to the sea water of a relatively new period such as Tertiary, when almost all of the petroleum deposits in Japan were formed.

From what has been stated above about the ancient sea water, the authors are convinced that the Japanese brine at least would not be the same as the sea water in its origin, but something other as it will be discussed in the following.

Existing Theories of the Origin of Petroleum in Japan. Now it is a matter of great importance to look upon the theories hitherto published on the origin of petroleum in Japan which must be closely related to the origin of brine. Among the theories of the origin of Japanese petroleum the two theories are commonly accepted, one being proposed by Takahashi[19] the other by Kobayashi.[20] According to Takahashi the organogenetic rocks which are found at oil strata in Japan may be the origin of petroleum because they always contain a minute quantity of kerosene often with a plenty of fossils of planktons, fishes and seaweeds, and these occurrence gave rôle of the formation of the brine water from the sea water whose composition differed little from the contemporary sea water. Although his idea for the brine water seems to surpass any other preceding theories in probable modification of the connate water theory instead of others' assuming the gradual transformation of the sea water into the brine with somewhat ambiguous changes, the brine as he considers is that which contains a large amount of alkaline earth elements such as the calcium and magnesium, whose phenomenon however is not the case in the Japanese brine as seen from Table 1. Kobayashi insisted on the so-called fish oil theory of the origin of the Japanese petroleum from the experimental facts of his own and coworkers i.e. fish oils yield petroleum-like hydrocarbons on dry distillation with Japanese acid clay at relatively low temperature and pressure. His theory is favorably accepted for the reason (1) that the distribution of the oil fields in Japan superposes approximately on that of the places producing acid clay and (2) that the oil fields can be considered to have situated at the seasides of the old times, where fishes were abundant. That the petroleum may be yielded from fish oils by the action of the acid clay only is not fully recognized, but that fish oils may be attributed to its origin is of some truth. In view of geological evidence most of Northern Honshiu of Japan was yet unseen on the sea in the early Tertiary age and fishes propagated in abundance owing to hotter climate than that of to-day as it is easily imagined, when occurred subsequent revolutions on land and sea which caused fishes to gather[21] near the seaside and to be buried under ground resulting in the formation of petroleum.

Grün and Wirth[22] who also supported the fish oil theory came to the conclusion that among fishes elasmobranchs are most probable as its origin because of the facts (1) that the livers of these fishes are commonly large as compared with their bodies and contain a great amount, 70–90% of the total, of a higher terpentine-like hydrocarbon "squalene" named by Tsujimoto,[23] (2) that this yields hydrocarbons of great resemblance to natural petroleum by thermal decomposition and (3) that elasmobranchs were prosperous in the age of petroleum formation.

Independent of these investigators Tanaka and Kuwata[24] separated, with naphthenic acid, from the petroleum from Ishikari in Hokkaido, Niigata and Akita Prefectures some saturated fatty acids such as palmitic, myristic, stearic, and arachidic among which the first acid being predominant, and found that the relative amounts of these fatty acids are almost identical with those in whale oil, shark liver oil, herring oil, or cod oil. They claimed from these facts that the animals which are most probable as the origin of petroleum may be the shark or whale in consideration of their inhabitation.

To these concordant results obtained by different investigators which lay stress on the fish especially of elasmobranchs oil theory the present authors will add an important suggestion resulting directly from the analyses of the Japanese brine from oil fields.

Similarity in the Compositions of the Brine and of the Fish Blood. The great resemblance between the composition of the Japanese brine and that of the bloods of fishes especially of elasmobranchs may be reasonably understood if we assume that the brine from an oil field comes from neither the sea water nor any underground water as the existing theories do, but from the body fluids of abundant corpses of fishes especially of elasmobranchs which are squeezed out by the compressive action of the earth crust, whose oily part being converted into petroleum at the same time.

The fact that the lack of the sulphate in the brine is simply explained by assuming as its origin elasmobranchs whose blood is lacking in the sulphate is of interest when we refer it to the rôle of sulphur compounds in the metabolism in the blood of higher animals. Indeed, elasmobranchs appeared already in Devon or Carboniferous ages while the other sea fishes in Jurassic and later ages.

According to Abderhalden[16] the inorganic composition of blood serum or plasma is not identical with that of blood corpuscle in the case of a higher animal such as the horse, pig, rabbit and man, and therefore it makes no estimation of the total inorganic blood composition from that of blood serum or plasma only. The blood corpuscles of these animals contain more potassium but less sodium than the blood sera or plasmas which are shown in Table 3. As for a sea animal Macallum(14) found that the total blood of the whale contains much potassium as the horse or pig does. Other higher animals such as the dog, cat, sheep, ox, deer, and etc., however, have no remarkable differences in the compositions of sera (or plasmas) and corpuscles, and such tendency would be the less, the lower an animal such as fish is. Smith's(15) results indicated on this point that the body fluids of fishes are very similar in inorganic composition with their blood sera or plasmas. From the above elucidation, the authors' hypothesis that the Japanese brine would come from the blood of fishes especially of elasmobranchs may be easily accepted, consistent with the fish oil theories of the origin of petroleum proposed by Tanaka and Kuwata and by Grün and Wirth. But the whale taken as the origin by the former authors must be rejected from the above Macallum's result.

As it has been often reported(25) that a comparatively large amount of iodine and bromine were found in the analyses of brine waters, the authors have determined these contents as well as the other constituents already shown. The result are shown in Table 4, in which the contents of these elements in the sea water observed by Fellenberg(26) and etc. are also shown. As seen from this table the molar ratios of bromine of the brine and of the sea water have no difference between them, while the iodine of the former is from 500 to 3000 times as much as that

Table 4.

Iodine and bromine contents of the Japanese brine and of the sea water.

	Iodine	Bromine	Authors
Brine from Innai, Akita Pref.	0.096	0.27	Ishikawa & Baba
Brine from Michikawa, Akita Pref.	0.015	0.16	,,
Brine from Niizu, Niigata Pref.	0.028	0.12	,,
Brine from Nishiyama, Niigata Pref.	0.037	0.26	,,
Brine from Ôtaki, Chiba Pref.	0.051	0.11	Ôhashi(6)
Sea water	0.000031	0.17 (Mean, from Tab. 2)	Fellenberg(26)

Table 5.

Iodine content of bloods of mammals.

Mammals	Iodine content×10^6 gr.%	Authors
Man	11.0	Turner[27]
,,	13	Kendall[27]
,,	8—12.8	Veil & Sturm[28]
Dog	11.1	Turner[27]
Sheep	4.1—1.5	Mauer & Ducrue[29]
Rabbit	3.1—0.7	,,

of the latter. The probable sources of such a considerable amount of iodine contained in the brines are perhaps due to marine lives, but the comparison of iodine content of the bloods of different animals with those of the brines can not unfortunately be done here because no appreciable data of fishes are found in the literature. In Table 5 the iodine content of a few mammals are given. Though somewhat low values are recorded in the cases of the sheep and rabbit, the iodine content of the bloods of mammals in general may be considered of the same order. Adopting the value 198 mg.% obtained by Kramer and Tisdall[18] as one of the probable values for the absolute sodium content in the human blood, the molar ratio of the iodine to the sodium is calculated to be 0.0010, which is much greater than that of the sea water (0.000031) but still smaller than that of brines (0.015–0.096).

Now, for fishes no such comparison can be made, but it is a well-known fact[30] that fishes especially elasmobranchs contain richer iodine than any other higher animals do, and it is also evident from the study of Mauer and Ducrue[29] that their bloods are rich in iodine when considered their foods. As regards the bromine content in the animal bloods no much mention is possible, but according to Bernhardt and Ucko[31] human blood contains from 1.0 to 1.5 mg.% in the normal state, which is equivalent to the molar ratio from 0.15 to 0.22, this indicating that the bromine content is perhaps universally constant in the animal bloods, the sea water and the brine.

If there is truth in what has been discussed for the origin of the brines, the brines from the same oil zone may have a constant or at least a similar composition with each other and conversely if underground water has a composition of the known brine type, it may give an oil indication together with a help of geological inquiry. On this hypothesis, the authors have examined mineral waters widely occurring in Japan whose analyses were completely carried out by the Imperial Hygienic Laboratories.(32) Among more than five hundred kinds of hot or cold mineral waters they have picked up three kinds of cold waters very similar to the brine type, whose composition is shown in Table 6 in the same expressions as before: The first is at Nagano in Niigata Prefecture, situated near Ômo, a famous oil field in the same Prefecture, the second is at Onishika, situated within the circumscription of the well-known Rumoi oil field in Teshio of Hokkaido. These two must of course be included in the same type of the brines shown in Table 1. The third is at Kashio in Nagano Prefecture where there has had as yet no production of petroleum, but an oil indication is beyond question according to the generally accepted geological opinion that it is situated on the famous Itoigawa-Shizuoka fault line, along which may exist the elongation of the great petroleum zone of northern part of Japan.

Table 6.

Composition of mineral waters of the brine type in Japan.

Mineral waters from	Na	K	Ca	Mg	Cl	Br	I	SO_4
1 Nagano, Niigata Pref.	100	0.55	1.77	3.18	100.7	0.10	0.010	0.015
2 Onishika, Hokkaido	100	0.93	3.50	2.37	105.8	0.11	0.034	—
3 Kashio, Nagano Pref.	100	0.83	3.25	0.66	108.9	0.04	trace	trace

NOTE:
(1) Nagano, Morimachi-mura, Minamikambara-gun, Niigata Prefecture.
(2) Onishika-mura, Rumoi-gun, Teshio Province, Hokkaido.
(3) Kashio, Oshika-mura, Shimoina-gun, Nagano Prefecture.

Conclusion. From the above results the authors have come to the conclusion that the Japanese brine from oil fields have a certain type of composition which is remarkably similar to the inorganic blood composition of fishes, especially of elasmobranchs and that this fact can be reasonably explained by assuming that the origin of the brine from oil fields are the body fluids of these fishes whose oils have been known to be most plausible as the origin of petroleum. Though the mechanism of the formation of petroleum as well as of brine water from the corpses of these fishes can not be fully accounted for, the above considerations will be of service to the investigation in the line.

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